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Formation of the maze domain structures in lithium niobate as a result of multiple pulse irradiation by infrared laser

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Abstract. The formation of the maze domain structures in the plates of congruent lithium niobate single crystal caused by multiple irradiation of infrared pulsed laser at different plate temperatures was studied. Four stages of domain evolution with increasing pulse number were distinguished. The dependence of the formed self-organizing domain structures on pulse number and sample temperature was revealed. Suppression of the domain formation at the elevated temperatures was attributed to increasing ionic conductivity, which led to decreasing the switching field.

1. Introduction

The modern development of telecommunication technologies requires manufacturing submicron and nanoscale periodic domain structures for use in various types of optical components, such as electrically controlled Bragg reflectors, beam control devices, and narrow-band filters [1-3].

Backward second harmonic generation (SHG), corresponding to the propagation of the wave with the fundamental frequency in the direction opposite to its second harmonic, requires the creation of submicron gratings with nanoscale accuracy of period reproduction [4]. Lithium niobate LiNbO_3 (LN) crystals being a favourite ferroelectric material for domain engineering [5] is considered as one of the best candidates for nanodomain structuring. Periodically poled LN crystals are used for converting the light frequency with record efficiency based on the quasi-phasematching effect [4,6-9]. The production of tailored domain structures is usually carried out by applying an electric field using photolithographically produced electrode patterns [10]. This method is ineffective for precise domain structuring with period below $2\text{ }\mu\text{m}$ [11]. The formation of stable nanodomain structures in congruent LN (CLN) crystals was demonstrated under highly non-equilibrium switching conditions [12-13]. Recently, the surface domain inversion leading to formation of the nanodomain structures has been proposed. The intense irradiation of the single domain LN by ultraviolet pulse laser induces formation of the shallow surface nanodomain rays with width below 100 nm and depth about $2\text{ }\mu\text{m}$ [14]. In contrast, the infrared pulsed laser irradiation forms the bulk domain structure of nanodomain rays with a depth of up to $200\text{ }\mu\text{m}$, which is enough for all above discussed applications [15]. It was shown experimentally that the polarization reversal occurred during cooling after termination of the laser heating.

In this paper, we show how multiple laser irradiation with controlled initial temperature affects morphology of the domain structure.



2. Experimental

The studied samples represented optically polished 0.5-mm-thick Z-cut plates of CLN (SIPAT Co., China). The pulsed CO₂ laser ($\lambda = 10.6 \mu\text{m}$, 40 W) with Gaussian distribution of laser energy density ($\omega = 0.35 \text{ mm}$) was used for the sample irradiation by pulses with duration (t_p) 3 ms and energy 160 mJ. The chosen duration of the pulse period (20 s) was enough for sample cooling to the initial temperature (T_i). Each series of pulse irradiation was carried out at fixed T_i (25, 50, 75, and 100°C). The radiation was focused on Z+ polar surface of the sample by ZnSe lens with focal distance of 50.8 mm. The power density was tuned with variation of the spot diameter by change of the distance between lens and irradiated surface.

The formed static domain structures were revealed by shallow selective chemical etching in pure HF for 13 seconds to prevent rearrangement of domain structure [16]. The surface relief corresponding to the domain structure was imaged by optical microscope Olympus BX51 (Olympus, Japan) [17].

3. Results and discussion

It was shown that the geometry of the domain structure formed after irradiation of CLN by series of laser pulses essentially changed with pulse number. The study of domains formed at different numbers of pulses at fixed temperature (25°C) allowed us to reveal four stages of domain formation. The first stage represented formation of the domain rays mostly oriented along Y crystallographic axis after the termination of the first pulse (Fig. 1a) [11]. During the second stage, with number of pulses below 20, the domain rays became wider with increasing number of pulses due to sideways domain wall motion (Fig. 1b). At the third stage, the flat domain walls became irregular with subsequent growth of the fingers on the wall for the number of pulses above 30 (Fig. 1c). At the last stage, for number of pulses above 70, the maze-like domain structure formed (Fig. 1d).

The statistical analysis of the domain images formed in the centre of irradiated zone allowed revealing the monotonous growth of the switched area with number of pulses (Fig. 2). The maximum fraction of the switched area, corresponding to the final domain structure, approached to 0.5. This is due to the fact that the total value of the pyroelectric field arising in periodically polarized ferroelectric crystal (e.g., of the Kittel type) is close to zero. For multiply irradiation at $T_i = 25^\circ\text{C}$, the fraction of the switched area stops changing after 80 pulses and the final domain structure appears (Fig. 1d).

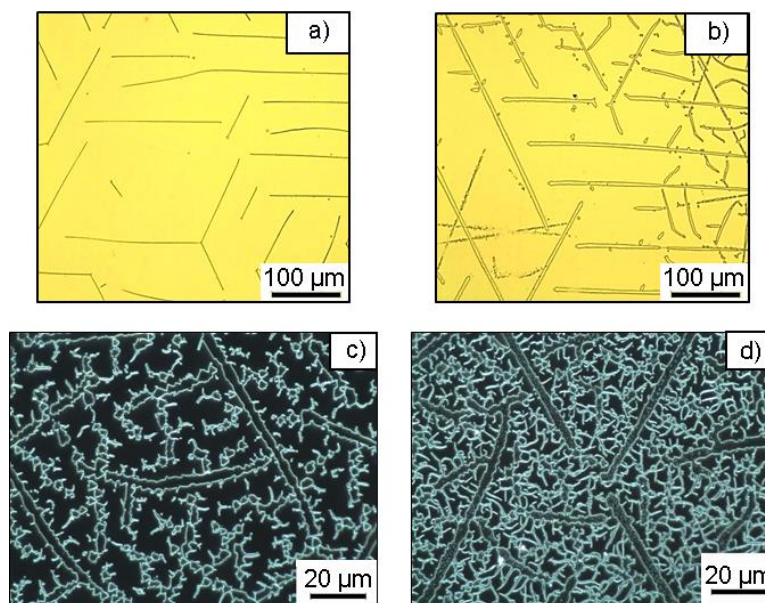


Figure 1. Optical visualization at (a, b) bright and (c, d) dark fields after chemical etching of domain structure after various number of pulses: (a) 1, (b) 20, (c) 50, and (d) 80.

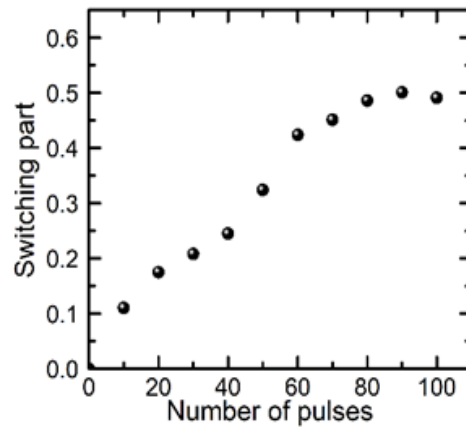


Figure 2. Dependence of the fraction of the switched area in the centre of irradiated zone on the number of pulses.

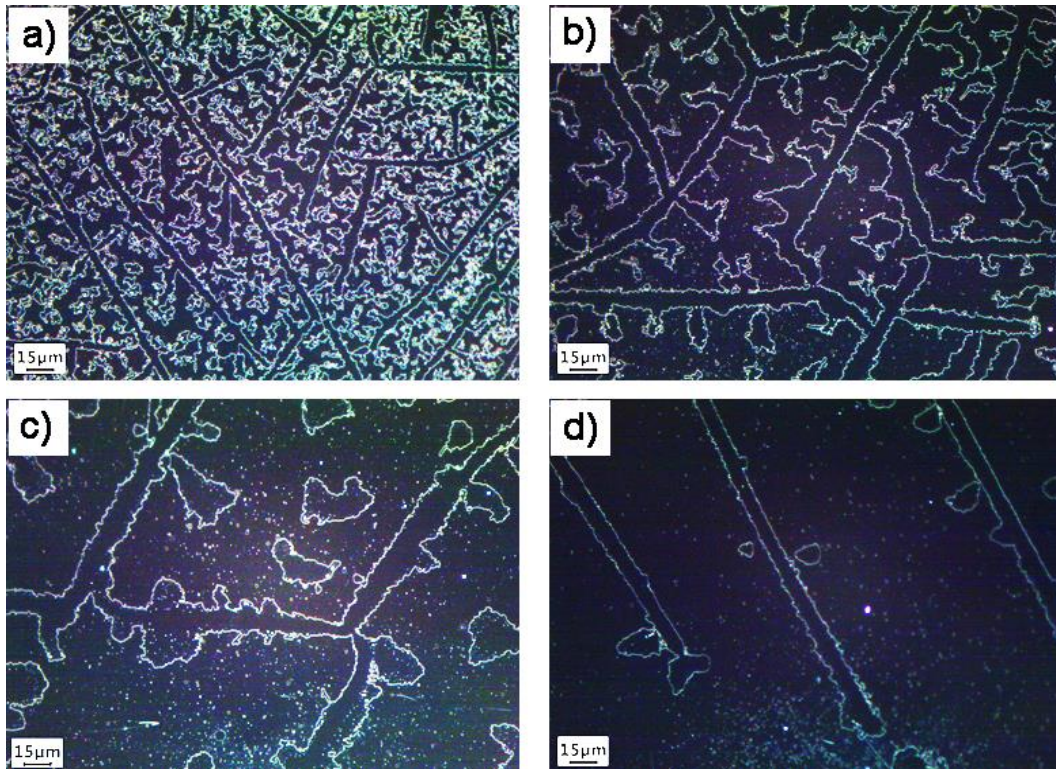


Figure 3. Domain structure after 100 pulses with initial temperature, °C: (a) 25, (b) 50, (c) 75, (d) 100.

It was obtained that the initial temperature of the CLN crystal plate strongly affected the final domain structure (Fig. 3). For quantitative characterization of the final domain structure at different T_i , two parameters were calculated: the fraction of the switched area and the domain wall density (total wall length per unit of the sample surface area). It was found that the fraction of the switched area linearly decreased with increasing T_i (Fig. 4a), whereas the domain wall density decreased rapidly with increasing temperature to 50°C and slowly decreased from 50 to 100°C.

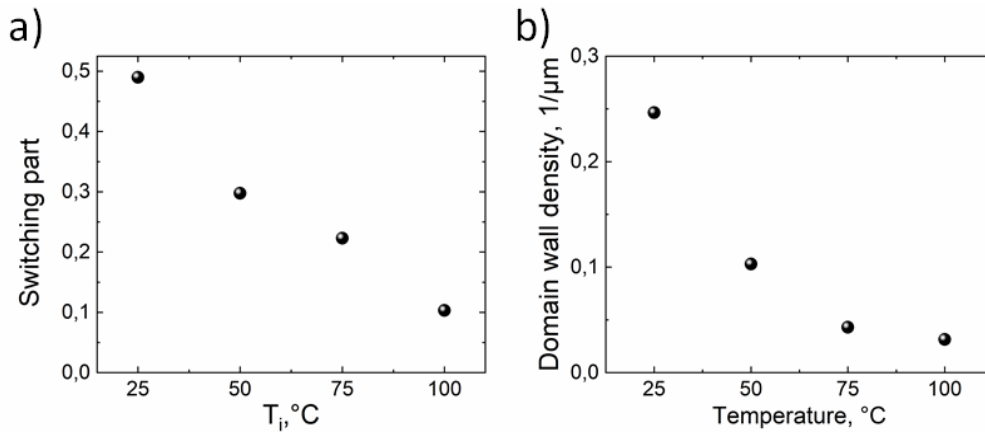


Figure 4. Dependence on T_i of: (a) the fraction of the switched area in the centre of the irradiated zone, (b) the average domain wall density.

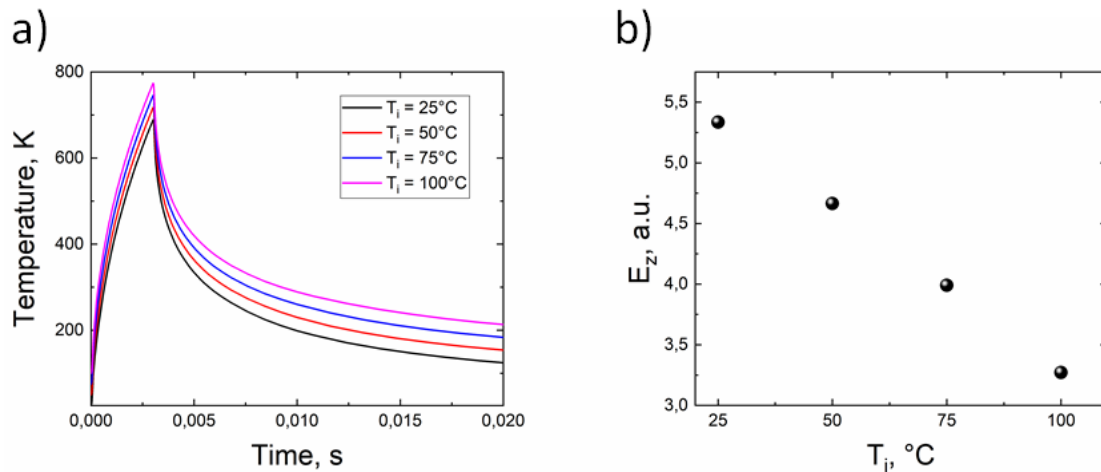


Figure 5. (a) The time dependences of the temperature in the centre of the irradiated zone for the first laser pulse for different T_i . (b) Dependence on T_i of calculated E_z values at 250°C during cooling.

For explanation of the obtained results, the time dependences of spatial distributions of temperatures and local pyroelectric fields in the bulk of CLN plate were calculated by Finite Element Method (FEM) using COMSOL Multiphysics software. The detail description of the used physical model was published in Ref. [19].

The time dependence of the temperature distribution during pulse laser heating for different T_i was calculated. The published temperature dependences of the heat capacity $c_p(T)$ and the thermal conductivity $k(T)$ for CLN were used [18]. The simulated time dependences of the temperature in the centre of irradiated zone for different T_i for the first laser pulse are presented in Figure 5a.

The time dependence of the pyroelectric field appeared as a result of temperature change was calculated. It was shown earlier experimentally that polarization switching started at 5-10 ms after heating termination [19], corresponding to 170-250°C. The calculated values of Z-component of the local field (E_z) at 250°C during cooling decreased with T_i (Fig. 5b). Decreasing of the switching field is due to increasing ionic conductivity of the crystal and leads to suppression of the domain formation.

4. Conclusion

The domain structure formation induced by multiple pulsed infrared laser irradiation in the plates of congruent lithium niobate was investigated. The dependence of the domain structure parameters on

number of pulses and initial temperature was measured. It was shown that number of laser pulses significantly affected the domain structure. Four stages of domain structure evolution were revealed: domain ray formation after the first pulse, domain widening by sideways domain wall motion, fingering, and formation of maze-like structure.

It was shown that increase in the initial plate temperature decreased the fraction of the switched area and the domain wall density. This fact can be attributed to decreasing the pyroelectric field due to growth of the ionic conductivity with initial temperature. It should be noted that the stable micro- and nanodomain structures obtained under the action of pyroelectric field attract interest for domain engineering.

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